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AGRICULTURAL AND HORTICULTURAL FUNGICIDAL COMPOSITION

Inventors:	Shigeru Hayashi 970-1 Shinya, Hamaoka-machi, Ogasa-gun, Shizuoka-ken
	Nobuyuki Yoshioka 27-10 Yanagigawa O-aza, Hasaki-machi, Kashima-gun, Ibaraki-ken
	Kanji Nakamura 203-29 Yamakiri, Shimizu-shi
	Yoshiaki Morimoto 1626 Gusagata, Shimizu-shi
Applicant:	Dainippon Ink and Chemical Inc. 35-58 Sakanoshita 3-chome, Itabashi-ku, Tokyo

Claims

1. An agricultural and horticultural fungicidal composition, which comprises tetrachloroisophthalonitrile and 1,17-diguanidino-9-azaheptadecane or salt thereof as effective components.

2. The agricultural and horticultural fungicidal composition according to Claim 1, wherein the mixing ratio of tetrachloroisophthalonitrile and 1,17-diguanidino-9-azaheptadecane or salts thereof is in the range of 1 to 20:1 by weight.

Detailed explanation of the invention

The present invention relates to an agricultural and horticultural fungicidal composition, and has the objective of offering a chemical agent that economically and effectively prevents pestilence.

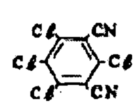
In recent years, environmental contamination due to chemical agents has been revealed as a serious problem, and there is a desire for minimizing the amount of chemical agents dumped into the environment, as well as reducing their effects on the environment.

Tetrachloroisophthalonitrile is currently an important agricultural and horticultural fungicidal composition that is used for a large number of vegetable and fruit diseases. However, tetrachloroisophthalonitrile is used in high concentrations of about 940-1500 ppm, which is a disadvantage.

The inventors of the present invention, et al., carried out selective investigations concerning chemical agents having synergistic action with respect to tetrachloroisophthalonitrile, and arrived at the present invention upon discovering that extremely high fungicidal action can be obtained by adding a small quantity of 1,17-diguanidino-9-azaheptadecane or salts thereof, which have potent chemotoxicity with respect to various types of crops at high concentrations.

Specifically, the agricultural and horticultural fungicidal composition of the present invention comprises tetrachloroisophthalonitrile and 1,17-diguanidino-9-azaheptadecane or salts thereof as effective components, and is formed from these substances together with various auxiliary agents.

Tetrachloroisophthalonitrile is represented by the formula:



In addition, 1,17-diguanidino-9-azaheptadecane is represented by the formula:

$$H_2N-\overset{\overset{NH}{\|}}{C}-NH(CH_2)_8NH(CH_2)_8NH-\overset{\overset{NH}{\|}}{C}-NH_2$$

This compound manifests excellent fungicidal effects on the level of those of tetrachloroisophthalonitrile. When allowed to stand, the compound reacts with atmospheric carbon dioxide gas to form carbonate, and in order to maintain it in stable form, the compound can be converted into stable salts having similar potency, or can be used unmodified. Examples of these types of salts that may be cited include hydrochloride, sulfate, acetate, citrate and p-toluenesulfonate. This 1,17-diguanidino-9-azaheptadecane is a well-known compound which, for example, can be synthesized following the method described in Japanese Kokoku Patent No. Sho 42[1967]-16607.

The mixing ratio of tetrachloroisophthalonitrile and 1,17-diguanidino-9-azaheptadecane or salts thereof must be selected so that the chemotoxicity of the 1,17-diguanidino-9-azaheptadecane is suppressed, while allowing sufficient manifestation of the effects of the tetrachloroisophthalonitrile at low concentration. Such ratios are in the range of 1-20:1 by weight, with preferred ratios being 1 [part] of the latter to 5-10 [parts] of the former.

Examples of the various types of auxiliary agents that may be cited include talc, bentonite, kaolin, clay, diatomaceous earth, white carbon and other solid carriers, ethanol, acetone and other solvents, polyoxyethylene sorbitan fatty acid esters, polyoxyethylene glycol ethers, polyoxyethylene alkyl ethers and other surfactants, and carboxymethylcellulose, gum arabic and other binders.

At the time of formulation, the required auxiliary agents are added, mixed or dissolved, and after uniform dispersion, the materials are processed into the desired shape in accordance with common methods.

By combining the tetrachloroisophthalonitrile and 1,17-diguanidino-9-azaheptadecane, good effects can be obtained against downy mildew and powdery mildew at low concentrations at which either component alone cannot provide protection, and at which there is no chemotoxicity with respect to plants.

In addition, the agricultural and horticultural fungicidal composition of the present invention has superior protective effects, not only as a protective agent against downy mildew and powdery mildew of cucurbitaceous plants, but also as a protective agent against gray mold, blight, anthracnose and other diseases in various types of plants.

The present invention is described in additional detail below using application examples, but there is a broad range of modifications allowable with respect to the binding agents and auxiliary agents that are admixed. The terms "parts" in the application examples denotes parts by weight.

Application Example 1 (powder)

Tetrachloroisophthalonitrile	5 parts
1,17-Diguanidino-9-azaheptadecane	1 part
White carbon	5 parts
Clay	89 parts

After mixing these components, the material was finely milled to produce a mixed powder.

Application Example 2 (hydrate)

Tetrachloroisophthalonitrile	50 parts
1,17-Diguanidino-9-azaheptadecane	5 parts
Polyoxyethylene nonyl phenyl ether	5 parts
Polyoxyethylene sorbitan monostearate	5 parts
Diatomaceous earth	35 parts

After mixing these components, the material was finely milled to produce a hydrate.

Application Example 3 (suspension)

Tetrachloroisophthalonitrile	20 parts
1,17-Diguanidino-9-azaheptadecane	5 parts
Polyoxyethylene styryl phenyl ether	3 parts
Ethylene glycol	15 parts
Water	57 parts

After mixing these components, the material was milled in a wet powdering device to obtain a suspension.

Application Example 4 (hydrate)

Tetrachloroisophthalonitrile	50 parts
1,17-Diguanidino-9-azaheptadecane	5 parts
Newcol 566	2 parts
Carplex	2 parts
Clay	41 parts

After mixing these components, the material was finely milled to obtain a hydrate.

The effects of the present invention are described below by providing test examples.

Test Example 1 (Downy mildew preventative effect)

Cucumber seeds (variety: Sagami Hanjiro) were planted in a 9-cm-square plastic box, and at the cotyledon stage, a conidiospore suspension of downy mildew (spore number: 1 x $10^{\text{[illegible]}}$/mL) was sprayed with a spray gun in the amount of 2 mL per box. The plastic box was placed in a 21°C humidification chamber, and after 24h, the hydrate prepared in Application Example 2 was diluted with water and applied in the amount of 30 mL per three boxes. Subsequently, the boxes were placed in the humidification chamber, and evaluation was performed according to the following criteria at 7 days after application. The infectivity was calculated according to the formula below.

Evaluation criteria:

n_0: Number of leaves on which no infection was observed.

n_1 Number of leaves in which the infected surface area was less than 1/3 the leaf surface area.

n_2 Number of leaves in which the infected surface area was at least 1/3, but less than 2/3, the leaf surface area.

n_3 Number of leaves in which the infected surface area was at least 2/3 the leaf surface area.

Infectivity (%): ($1n_1 + 2n_2 + 3n_3$/total leaf surface area x 3) x 100

The results are presented in Table 1.

Table 1

1 実験No.	有効成分濃度 (ppm) 2		発病度 (%) 5
	テトラクロロイソフタロニトリル 3	1,17-ジグアニジノ-9-アザヘプタデカン 4	
1	500	0	100
2	0	50	68.9
3	500	50	8.6
4	250	50	21.2
5	0	0	100

Key: 1 Experiment No.
2 Effective component concentration (ppm)
3 Tetrachloroisophthalonitrile
4 1,17-Diguanidino-9-azaheptadecane
5 Infectivity (%)

Test Example 2 (Powdery mildew protective effect)

Cucumber seeds (type: Sagami Hanjiro) were planted in a 9-cm-square plastic box, and at the cotyledon stage, the hydrate prepared in Application Example 2 was diluted with water, and applied in the amount of 30 mL per three boxes using a spray gun. The boxes were then placed in a humidification chamber, cucumber seedlings infected with powdery mildew were placed in the vicinity, and natural infection was allowed to occur. 14 days after application, investigations were carried out based on the following criteria, and the infectivity was calculated according to the formula below.

Evaluation criteria:

n_0: Number of leaves on which no infection was observed.

n_1 Number of leaves in which the infected surface area was less than 1/3 the leaf surface area.

n_2 Number of leaves in which the infected surface area was at least 1/3, but less than 2/3, the leaf surface area.

n_3 Number of leaves in which the infected surface area was at least 2/3 the leaf surface area.

Infectivity (%): $(1n_1 + 2n_2 + 3n_3/\text{total leaf surface area} \times 3) \times 100$

The results are presented in Table 2.

Table 2

1 実験No	有効成分濃度（ppm） 2		発病度（%） 5
	テトラクロロイソフタロニトリル 3	1,17−ジグアニジノ−9−アザヘプタデカン 4	
6	250	0	85.6
7	0	50	52.3
8	250	50	10.3
9	250	25	18.6
10	0	0	100

Key:
1 Experiment No.
2 Effective component concentration (ppm)
3 Tetrachloroisophthalonitrile
4 1,17-Diguanidino-9-azaheptadecane
5 Infectivity (%)

Test Example 3

The hydrate prepared in Application Example 4 was diluted with water, and applied a total of 4 times every 7 days onto cucumber (variety: Shintokiwa) in 1/5000 are Wagner pots with 2-3 open leaves per plant. The Wagner pots were placed in a humidification chamber after the first chemical application, and cucumber seedlings infected with powdery mildew were placed in their vicinity in order to allow natural infection. Infectivity at 28 days after the first chemical application is presented in Table 3. Infectivity was calculated in the same manner as in Application Example 1.

Table 3

1 実験№	有効成分濃度 (ppm) 2		5 罹病度 (%)
	テトラクロロイソフタロニトリル 3	1,17-ジグアニジノ-9-アザヘプタデカン 4	
11	250	0	84
12	125	0	64
13	62.5	0	65
14	0	50	19
15	0	25	23
16	0	125	26
17	250	50	0
18	125	25	0
19	62.5	12.5	7
20	0	0	85

Key: 1 Experiment No.
2 Effective component concentration (ppm)
3 Tetrachloroisophthalonitrile
4 1,17-Diguanidino-9-azaheptadecane
5 Infectivity (%)

Test Example 4 (Yoshiba Helminthosporium leaf blight protective effect)

Shibafu (pentarosu [transliteration] bent grass) was sectioned into 25 x 30-cm areas, and the hydrate prepared in Application Example 4 was diluted with water, before applying at a rate of 1 g/L/m^2. After drying, a suspension of Yoshiba leaf blight spores (200 per field of view at 100X) was applied by spraying at a rate of 0.1 L/m^2, and the grass was left in a 25°C humidification chamber for 3 days.

The results are presented in Table 4.

Table 4

1 実験No	有効成分濃度 (ppm) 2		被害面積率(%) 5
	テトラクロロイソフタロニトリル 3	1，17－ジグアニジノ－9－アザヘプタデカン 4	
21	250	0	35
22	125	0	40
23	62.5	0	50
24	0	50	30
25	0	25	45
26	0	12.5	70
27	250	50	1
28	125	25	5
29	62.5	12.5	10
30	0	0	85

Key: 1 Experiment No.
2 Effective component concentration (ppm)
3 Tetrachloroisophthalonitrile
4 1,17-Diguanidino-9-azaheptadecane
5 Infectivity (%)

Test Example 5 (Powdery mildew protective effect)

Cucumber plants (variety: Shintokiwa) planted in 1/5000 are Wagner pots having 2-3 opened leaves per plant were treated by a total of four applications at intervals of 7 days using chemical liquid produced by blending the prescribed amount of 17-diguanidino-9-azaheptadecane with tetrachloroisophthalonitrile. The Wagner pots were placed in a greenhouse after the first application, cucumber seedlings that had been infected with powder mildew were placed in the vicinity thereof, and infection was allowed to occur. The infectivity was calculated using the same criteria as in Test Example 1 on the day after the fourth application of the chemical agent. The chemotoxicity was denoted by "+++" for extreme, "++" for pronounced, "+" for moderate, "±" for slight, and "-" for nonexistent. The results are presented in Table 5

Table 5

1 実験No.	2 有効成分濃度 (ppm)		5 配合比 A:B	6 発病度 (%)	7 薬害の程度
	3 テトラクロロイソフタロニトリル(A)	4 1,17-ジグアニジノ-9-アザヘプタデカン(B)			
1	100	500	1:5	0	卌
2	250	250	1:1	0	±
3	500	25	20:1	15	−
4	500	125	50:1	35	−
5	500	50	10:1	9	−
6	500	100	5:1	0	−
7	0	0	−	95	−

Key:
1 Experiment No.
2 Effective component concentration (ppm)
3 Tetrachloroisophthalonitrile (A)
4 1,17-Diguanidino-9-azaheptadecane (B)
5 Blending ratio A:B
6 Infectivity (%)
7 Chemotoxicity level